# HYDROGEN EFFECTS ON LOCALIZED PLASTICITY IN A METALLIC COMPOSITE

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It is experimentally shown that macroscopic plastic flow is localized in a layered composite produced via twopass vacuum electron beam cladding. The quantitative regularities of macroscopic localization of plastic deformation in the base and cladded layers of A570-36/A516-55 composite are compared at different stages of work hardening before and after electrolytic hydrogen saturation. The main characteristics of localized plastic flow under these conditions are determined via double-exposure speckle photography combined with uniaxial tensile testing. It is found that hydrogen saturation within 24 h leads to the enhancement of localized plastic deformation.

Keywords: layered metallic composites, plastic flow localization, hydrogen embrittlement

## INTRODUCTION

Despite the long-term study of hydrogen, there is still no unified point of view on the mechanisms of its influence on the metal destruction. This is due to a complex of interrelated factors among which are structure, phase composition, type of crystal lattice, test temperature, etc. In works [1–4], the main hypotheses of hydrogen embrittlement (HE) of structural materials used in water-cooling environments (such as hydrogen-induced decohesion, hydrogen-formed brittle pseudo-hydrides, and localized and hydrogen-enhanced plasticity) have been considered by the examples of steels of various compositions. Special attention has been paid to the effect of hydrogen on the interatomic bonds, the possible correlation between hydrogen-induced phases and HE, and hydrogen-induced early microplastic deformation.

In work [5], the synergistic effects of the deformation response in thermodynamically open metal-hydrogen systems were considered. The authors noted that this response was observed under certain conditions, especially when the hydrogen diffusion coefficient in the material was  $\sim 10^{-9}-10^{-10}$  m<sup>2</sup>/s and the hydride (vanadium, tantalum, or palladium) or hydride-like (iron) phases form according to the martensitic transition mechanism. The structure of the hydride phases in this case is due to the ordering of hydrogen atoms in the crystal lattice of the matrix with the formation of the hydrogen sublattice. Like the conventional thermoelastic transformations, the phase transition in the above materials is preceded by a series of pre-transition states characterized by a *softening* of the shear modulus. In metal-hydrogen systems, the decrease in the shear modulus ultimately leads to the emergence of a particular *quasi-liquid* state with a relatively high resistance of the metal to normal stresses and a very low resistance to shear deformation.

Most theories and models of hydrogen brittleness use the critical hydrogen concentration, but neither analytical nor experimental methods for determining this value have been proposed. In work [6] it is postulated that the

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Material	С	Si	Mn	Ni	S	Р	Cr	Fe
A570-36	0.14-0.22	0.12-0.3	0.4-0.65	0.3	0.05	0.04	0.3	98
A 516-55	0.12	0.5-0.8	1.3-1.7	0.3	0.04	0.035	0.3	97

TABLE 1. Chemical Composition

combination of critical hydrogen concentrations and the stress coefficient at the crack tip is a constant of the material under consideration.

An experimental justification of the synergistic law and the damage parameter was obtained in work [7] based on the thermodynamic analysis of the nonequilibrium metal-hydrogen system. The proposed relationship allows one to determine the tear resistance and to compare it with the concentration of hydrogen in the rupture zone independently of the initial strength and plasticity of the material. At the same time, the state of hydrogen (volatile (diffusion-mobile) or molized (bound)) can be reliably distinguished. It is shown that the decrease in the tear resistance (the level of damage) of steel increases proportionally to the concentration of hydrogen [7]. This makes it possible to determine the parameter of hydrogen damage, which characterizes the tendency of steel to hydrogen brittleness and depends on the properties of the material.

Hydrogen is conducive to localized plastic flow [8, 9]; however, its effect is clear so far only at the microscale level [10, 11]. In work [12], the autowave theory of plasticity [13, 14] has been explained and applied to describe the influence of hydrogen on the plasticity. The quantitative patterns of macroscopic localization of plastic deformation in ferritic and austenitic steels as well as in aluminum and titanium alloys were compared at different stages of strain hardening before and after electrolytic hydrogen saturation. The main characteristics of localized plastic flow under these conditions were determined using double-exposure speckle photography combined with mechanical tests [13]. In view of the above, this study aims at considering the impact of electrolytic hydrogen saturation on macroscopically localized plastic flow in a metal layered composite produced via electron beam cladding.

#### MATERIALS AND EXPERIMENTAL METHODS

In this work, the composite was a 5.5 mm thick substrate of structural steel A570-36, which was covered with a 3 mm thick layer via two-pass electron beam cladding of the A516-55 steel wire in vacuum conforming to the procedure described in work [15]. The chemical composition of the composite is given in Table 1. The substrate metal possessed a fine-grained ferrite-pearlite structure with the average ferrite grain size of ~18  $\mu$ m [15]. The grain refinement of the base substrate metal (A570-36) to 10–15  $\mu$ m was observed in the 0.2–0.4 mm wide transition zone toward the cladded layer (A516-55). In turn, the cladded layer exhibited a ferrite-pearlite structure with predominately ferrite grains (~40  $\mu$ m) [15].

The double-sided blade samples with dimensions of  $40 \times 8.5 \times 2$  mm were obtained via electrical discharge machining whereby the material was cut perpendicular to the substrate plane so that the displacement field distribution could be achieved during stretching in different layers of the lateral face of the composite. The uniaxial tensile tests were carried out using a Walter + Bai LFM-125 universal testing machine at room temperature with a constant speed of the movable grip of  $3.3 \cdot 10^{-6}$  m/s.

The mechanical properties and plastic deformation heterogeneity of the composite were investigated in the initial state (stage 1) and after 24 h of laboratory electrolytic hydrogen saturation of samples (stage 2). The samples prepared for uniaxial tensile testing were electrolytically saturated with hydrogen in a thermostated three-electrode electrochemical cell with a graphite anode at a constant cathode potential of -670 mV relative to the silver chloride reference electrode. The saturation was implemented in a 1 M sulfuric acid solution with the addition of 20 mg/l thiourea at a temperature of 323 K under preliminary purging with nitrogen [16]. The current-voltage curves were recorded by an IPC-Compact potentiostat. The hydrogen concentrations were estimated taking into account the conditions of potentiostatic electrolytic hydrogenation (cathode potential, temperature, and time of hydrogen saturation of the sample), sample thickness, and hydrogen diffusion coefficient [16].

	State	σ <sub>0·2</sub> , MPa	σ <sub>UTS</sub> , MPa	$\varepsilon_B, \%$	δ, %	ε <sub>loc</sub> , %	$\alpha = \varepsilon_{loc} / \delta$	$\beta = \varepsilon_{loc} / \varepsilon_B$
1	Substrate	$270 \pm 2$	401 ± 1.5	$35\pm2$	$38 \pm 1.8$	$20 \pm 1$	$0.53\pm0.09$	$0.57\pm0.08$
	Cladding layer					$17 \pm 1$	$0.44 \pm 0.04$	$0.48\pm0.06$
2	Substrate	191 ± 3	$280\pm2.3$	$10 \pm 1.5$	$14 \pm 2$	$8 \pm 1$	$0.57\pm0.06$	$0.8\pm0.05$
	Cladding layer					$5 \pm 1$	$0.36\pm0.05$	$0.5 \pm 0.03$

TABLE 2. Mechanical Characteristics of A570-36/A516-55 Composite



Fig. 1. Patterns of localized plastic flow in the base layer of the A570-36/A516-55 composite in state 1 (*a*) and state 2 (*b*) in the form of local elongation distributions  $\varepsilon_{xx}(x, t)$  at the yield point (the stationary grip is at the origin).

The mechanical tests of flat samples were combined with acquisition and analysis of localized plasticity patterns via double-exposure speckle photography [12–14]. This allowed one to restore the displacement vector fields and to calculate the plastic distortion tensor components, thereby obtaining information about plastic flow regularities. The details and possibilities of the technique are available in work [17].

#### EXPERIMENTAL RESULTS AND DISCUSSION

The stress-strain  $\sigma(\varepsilon)$  curves showed that the hydrogenation quantitatively changed the response of the material to deformation, but almost did not affect the shape of the curves themselves. In all cases, the yield point, parabolic hardening, and pre-fracture stages were clearly distinguished. The mechanical characteristics (yield strength  $\sigma_{0.2}$ , strength limits  $\sigma_{UTS}$ , total strain at the ultimate tensile strength  $\varepsilon_B$ , and relative elongation to failure  $\delta$ ) of the initial (state 1) and hydrogen-saturated (state 2) samples are shown in Table 2, revealing a sharp decrease in the strength and ductility of the material. In contrast to state 1, the failure of samples saturated with hydrogen occurred without visible neck formation. The coefficient of hydrogen embrittlement was determined as  $K_H = [(\delta_0 - \delta_H)/\delta_0] \times 100\% = 63\%$ , where  $\delta_0$  and  $\delta_H$  are the total elongations at fracture of unhydrogenated and hydrogen-induced microcracks. Their emergence in the plasticity and strength in the composite was associated with the hydrogen-induced microcracks. Their emergence of which makes the material damage after reaching critical values [18].

The local elongation distributions at the yield point of the A570-36 base layer (state 1) exhibited a single zone of localized plastic flow similar to the Chernov–Lüders band (CLB), which originated near the movable grip of the test machine and then passed from the substrate to the A516-55 cladded layer, moving to the stationary grip at a speed of  $0.8 \cdot 10^{-3}$  m/s (Fig. 1*a*).

After 24 h of electrolytic saturation with hydrogen (state 2), the local deformation distributions at the yield point (Fig. 1*b*) in the base and cladded layers demonstrated two zones of localized deformation (CLB), which moved toward each other at the speeds of  $2.3 \cdot 10^{-4}$  m/s and  $5.0 \cdot 10^{-5}$  m/s. The *meeting* of the described deformation foci corresponded to the end of the yield point stage.



Fig. 2. Strain maps in the form of total local elongation isolines  $\varepsilon_{xx}(x, \varepsilon)$  along the tension axis X with increasing total deformation  $\varepsilon$  of the A570-36/A516-55 composite for (a) the base and (b) cladded layers in the state (1).

In states 1 and 2, a system of stationary equidistant zones of plastic deformation localization was detected in the base and cladded layers of the composite at the stage of parabolic hardening. It was established that hydrogenation led to a decrease in the spatial period of localized plasticity autowaves from 6.5 mm for state 1 to 5.0 mm for state 2. Further, the system of stationary foci of localized plastic deformation, a characteristic of the parabolic hardening stage, was replaced by a gradually growing stationary maximum with the large amplitude of the local elongation component of the plastic distortion tensor  $\varepsilon_{xx}$ . This maximum indicates the location of the future viscous fracture of the samples in states 1 and 2, and since its appearance, the deformation is almost entirely localized in this narrow zone of the material. At the final stage, the main crack in the composite originated in the joint area from the base layer in state 1 and the cladded layer in state 2, and then spread through the whole cross section of the sample.

The sequential summation of local deformations in the localized plasticity zones with an increase in total deformation  $\varepsilon$  allows one to identify the fracture center at the stages preceding the deformation at the ultimate strength  $\varepsilon < \varepsilon_{\rm B}$ . An example of such a summation of local elongations  $\varepsilon_{xx}$  for the middle line *X* of the base and cladded layers of the composite is shown in Figs. 2 and 3 in the form of isolines.

It can be seen that for a fixed value of the total deformation, the frequency and curvature of the isolines vary for different layers of the composite and for states 1 and 2. In the cladded layer of the composite in state 1, the total deformation at the stage of parabolic hardening  $\varepsilon_{loc} = 17$  % was mainly localized in a stable zone with a coordinate X = 15 mm (Fig. 2*b*), which subsequently corresponded to the neck formation site at  $\varepsilon_B = 35$  % and the rupture of the sample at  $\delta = 35$  %. In the deposited layer of the composite in state 2, the total deformation at the stage of parabolic hardening  $\varepsilon_{loc} = 5$  % was detected in the zone near X = 30 mm (Fig. 3*b*), which also corresponded to the neck formation area at  $\varepsilon_B = 10$  % and the rupture of the sample at  $\delta = 14$  % (Table 2). Electrolytic hydrogen saturation of the base and cladded layers enhanced the localized plasticity and, consequently, the isoline intensities (Fig. 3). Therefore, these experimental results provide the complementary information about localized plasticity processes in layered composites and open up the possibility of predicting the time and location of the fracture site before the material undergoes the tensile strength and the fracture neck is formed.

Table 2 displays the values of deformations  $\varepsilon_{loc}$  which refer to the visualized stable zones of localized plastic deformation for the base and cladded layers in initial state 1 and after saturation with hydrogen (state 2). During hydrogen embrittlement, there is not only a decrease in plasticity, but also in the duration of pre-failure focus nucleation. While the ratio of deformations  $\alpha = \varepsilon_{loc}/\delta$  reveals the proportion of homogeneous deformation from the total deformation of the material to failure, the ratio  $\beta = \varepsilon_{loc}/\varepsilon_B$  indicates the contribution of homogeneous deformation before



Fig. 3. Strain maps in the form of total local elongation isolines  $\varepsilon_{xx}(x, \varepsilon)$  along the tension axis X with increasing total deformation  $\varepsilon$  of the A570-36/A516-55 composite for (*a*) the base and (*b*) cladded layers in the hydrogenated state (2).

the neck formation [19]. According to Table 2, the values of the parameters  $\alpha$  and  $\beta$  were different for various layers of the composite in the initial and hydrogenated states.

Within the framework of the autowave model of localized plasticity [13], the emergence of a stable localization zone is associated with the onset of the collapse of the localized plasticity autowave, that is, with the contraction of the autowave into the fracture neck of the metal or alloy. In this work, the autowave collapse of localized plastic flow was characterized by the deformation  $\varepsilon_{loc}$  and the parameter  $\beta$ . In initial state 1, the collapse was detected much later than after saturation with hydrogen (state 2). Therefore, when a composite part is used in a hydrogen-containing environment, its parameter can decrease by 60 % and the degradation of the material will occur faster.

#### CONCLUSIONS

It was shown that 24 h electrolytic hydrogen saturation of the A570-36/A516-55 layered composite obtained via two-pass vacuum electron beam cladding altered the duration of epy strain hardening stage and the relevant local deformation distributions in the base and cladded layers. Hydrogen enhanced the localized plastic flow at the macroscopic level, which manifested itself by the change in the spatial period of the local deformation distribution and the rate of propagation of deformation foci at different stages of strain hardening. The amount of deformation corresponding to the pre-failure focus nucleation during hydrogen saturation decreased during the parabolic hardening, reducing the stability of the above-mentioned stage. The possibility was shown as well of predicting the total deformation and the coordinates of the pre-fracture focus before the onset of the tensile strength and the formation of the fracture neck in a layered composite.

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